

# Thermopower across the pseudogap critical point of $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ : Evidence for a quantum critical point in a hole-doped high- $T_c$ superconductor

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The thermopower  $S$  of the high- $T_c$  superconductor  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  was measured as a function of temperature  $T$  near its pseudogap critical point, the critical hole doping  $p^*$  where the pseudogap temperature  $T^*$  goes to zero. Just above  $p^*$ ,  $S/T$  varies as  $\ln(1/T)$  over a decade of temperature. Below  $p^*$ ,  $S/T$  undergoes a large increase below  $T^*$ . As with the temperature dependence of the resistivity, which is linear just above  $p^*$  and undergoes a large upturn below  $T^*$ , these are typical signatures of a quantum phase transition. This suggests that  $p^*$  is a quantum critical point below which some order sets in, causing a reconstruction of the Fermi surface, whose fluctuations are presumably responsible for the linear- $T$  resistivity and logarithmic thermopower. We discuss the possibility that this order is the “stripe” order known to exist in this material.

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The nature of the pseudogap phase in high- $T_c$  superconductors has yet to be elucidated. Quantum oscillation studies [1] have revealed that the large hole-like Fermi surface characteristic of highly overdoped cuprates [2] is modified in the pseudogap phase, where it contains small electron-like pockets [3]. A fundamental question is: what causes this change in Fermi surface? Is it the onset of some order? If so, what symmetry is broken?

Recent measurements of the normal-state resistivity  $\rho(T)$  and Hall coefficient  $R_H(T)$  in the hole-doped cuprate  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  (Nd-LSCO) have shown that the change in Fermi surface in this material occurs at the critical doping  $p^*$  where the pseudogap temperature  $T^*$  goes to zero [4], as illustrated in Fig. 1. Recent photoemission measurements on Nd-LSCO at  $p = 0.12$  have confirmed that the pseudogap in Nd-LSCO has the same features as in other cuprates [5], with an onset temperature consistent with  $T^*$  determined from transport (see Fig. 1). At a hole doping  $p = 0.20$ ,  $\rho(T)$  shows a pronounced upturn below  $T^*$  (see inset to Fig. 1). The simultaneous upturn in  $R_H(T)$  is strong evidence that these upturns are caused by a change in the Fermi surface [4]. At a slightly higher doping,  $p = 0.24$ ,  $R_H(T)$  remains flat at low temperature (see inset in top panel of Fig. 2), with the value expected of a large hole-like Fermi surface containing  $1 + p$  holes [4]. In that large-Fermi-surface state,  $\rho(T)$  is linear in temperature down to the lowest temperatures [4] (see inset to Fig. 1).

In this Letter, we investigate the thermopower  $S(T)$  of Nd-LSCO. In general, the thermopower is a complex quantity that involves the energy dependence of the conductivity [9, 10]. However, in the limit of dominant impurity scattering, it has been shown theoretically that  $S/T \propto (C_e/T)(1/ne)$ , where  $C_e$  is the electronic specific heat,  $n$  is the density of charge carriers and  $e$  is the charge

of the electron [10]. Empirically, it has been pointed out that  $S/T \approx (C_e/T)(1/ne)$  in the limit of  $T \rightarrow 0$  for a wide range of strongly correlated electron systems [9]. Therefore, at low temperature the thermopower approximately represents the electronic heat capacity per charge carrier. (Note that it would be difficult to measure  $C_e(T)$  accurately in Nd-LSCO given that it is less than 1 % of the total specific heat  $C(T)$  above 4 K, and the low-temperature behavior is masked by a large Schottky anomaly [11, 12].) We find that the three regimes of behavior seen in the resistivity as upturn for  $p < p^*$ , linear for  $p = p^*$  and quadratic for  $p > p^*$ , show up in  $S/T$  respectively as upturn, logarithmic divergence and nearly flat. This is strongly reminiscent of the electron behavior in metals near a quantum phase transition [13], suggesting that the pseudogap phase is characterized by some order, which vanishes at a quantum critical point located inside the region of superconductivity in the phase diagram. Evidence points to so-called “stripe” order, as the anomalies in transport coincide with the onset of spin / charge modulations.

The two samples of Nd-LSCO used in this study are the same as used and described in Ref. [4]. They have a doping of  $p = 0.20$  and  $p = 0.24$ , with respective  $T_c$  values of 20 K and 17 K. The thermopower was measured using a one-heater, two-thermometer DC technique, with Cernox thermometers. The applied temperature gradient was always less than 7 % of the average sample temperature. The thermopower of the resistive leads in the measurement circuit (PtW or phosphor-bronze) was calibrated against optimally-doped  $\text{YBa}_2\text{Cu}_3\text{O}_y$  ( $T_c = 93$  K) for  $T < 90$  K and 6N-pure Pb for  $T > 90$  K [14]. The  $p = 0.24$  sample was also measured using a low-frequency two-heater, two-thermometer AC technique [15], with a sinusoidal excitation of frequency 5–100 mHz and ampli-

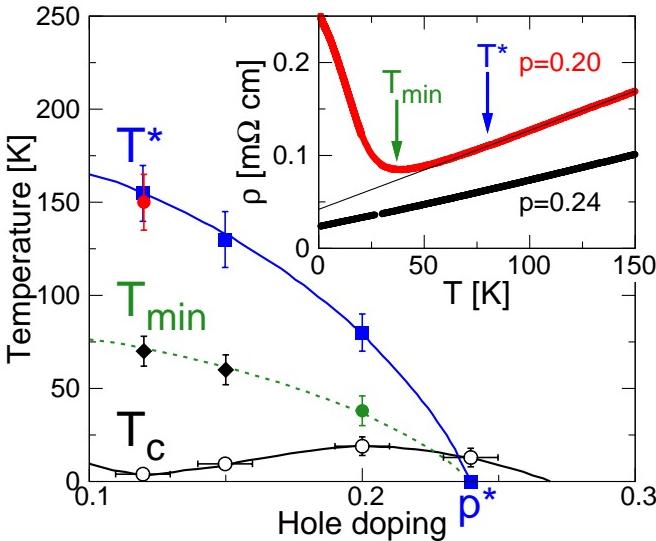


FIG. 1: (Color online) Phase diagram of Nd-LSCO. The pseudogap temperature  $T^*$  (blue squares and solid line) is defined as the temperature below which the normal-state resistivity deviates from its linear- $T$  behavior at high temperature (see inset and [4]). This is in agreement with the (extrapolated) opening of the pseudogap seen by photoemission at  $p = 0.12$  (red circle) [5]. Since the linear- $T$  dependence extends down to  $T \rightarrow 0$  at  $p = 0.24$  (see inset),  $T^* = 0$  at that doping. Although we define the pseudogap critical point  $p^*$  to be at  $p = 0.24$ , it could be slightly lower. The superconducting transition temperature  $T_c$  (open black circles) is also plotted, showing that the  $T^*$  line must end inside the superconducting phase. Data for  $p = 0.12$  and  $p = 0.15$  are from [6]; data for  $p = 0.20$  and  $p = 0.24$  are from [4]. Also shown is the upturn temperature  $T_{\min}$  (closed green circle and dashed line) at which the resistivity reaches its minimum value (see inset). The onset of charge order deduced from X-ray diffraction [7, 8] (black diamonds) coincides with  $T_{\min}$  (see Refs. [4, 6]). Inset: normal-state resistivity of the two Nd-LSCO crystals used in this study, measured in a magnetic field strong enough to entirely suppress superconductivity (from [4]).

tude 0.1 K. The signal-to-noise ratio in the AC measurement was 10 times better than in the DC case. There was excellent agreement in the data obtained with both techniques.

In Fig. 2, we plot  $S/T$  vs  $\log T$  for Nd-LSCO at  $p = 0.20$  and  $p = 0.24$ . The data taken on our  $p = 0.20$  crystal ( $x = 0.20$  and  $y = 0.4$ ) is in excellent quantitative agreement with previous measurements on  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  at the same values of  $x$  and  $y$ , over the entire temperature range in zero magnetic field [16]. To our knowledge, no in-field thermopower data has been reported on Nd-LSCO, nor is there any published data on Nd-LSCO for  $x > 0.20$ . Our data on Nd-LSCO at  $p = 0.24$  is in good quantitative agreement with published data on polycrystalline LSCO at  $p = 0.25$  (only reported in zero field) [17, 18].

There is no consensus on the mechanism that governs the thermopower in cuprates. While phonon drag has

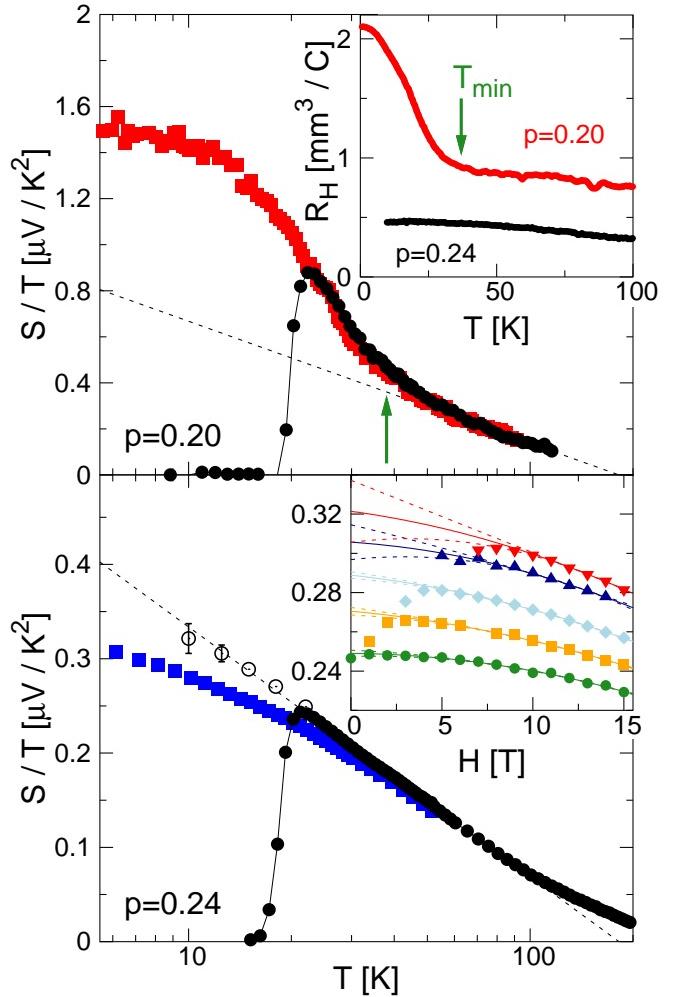


FIG. 2: (Color online) Thermopower  $S(T)$  of Nd-LSCO, plotted as  $S/T$  vs  $\log T$ , with (squares) and without (full circles) a magnetic field of 15 T applied along the  $c$ -axis. Top panel: sample with  $p = 0.20$ , measured with the DC technique (see text). The magnetic field has no discernible effect other than to suppress superconductivity. The dashed line is a linear fit to the data above 50 K. The arrow marks  $T_{\min}$  (see inset of Fig. 1). Inset: Hall coefficient as a function of temperature, for both Nd-LSCO samples [4]. At  $p = 0.20$ , the upturn in  $R_H(T)$  at low temperature is seen to coincide with  $T_{\min}$ . Bottom panel: sample with  $p = 0.24$ , measured with the AC technique (see text). The magnetic field is seen to cause a reduction of  $S$  for  $T < 40$  K. To correct for this and extend the zero-field behavior to  $T < T_c$ , we extrapolate finite field data to zero field (see inset). The resulting extrapolated values are plotted as open circles. Inset:  $S/T$  for Nd-LSCO with  $p = 0.24$  as a function of magnetic field, at five fixed temperatures: 10, 12.5, 15, 18 and 22 K (top to bottom). Second order polynomial fits to the field dependence are extrapolated back to zero field. Best (solid lines) and worst (dashed lines) fits are shown, indicative of the uncertainty in the width and position of the superconducting downturn. The corresponding error bars are shown in the main panel (open circles).

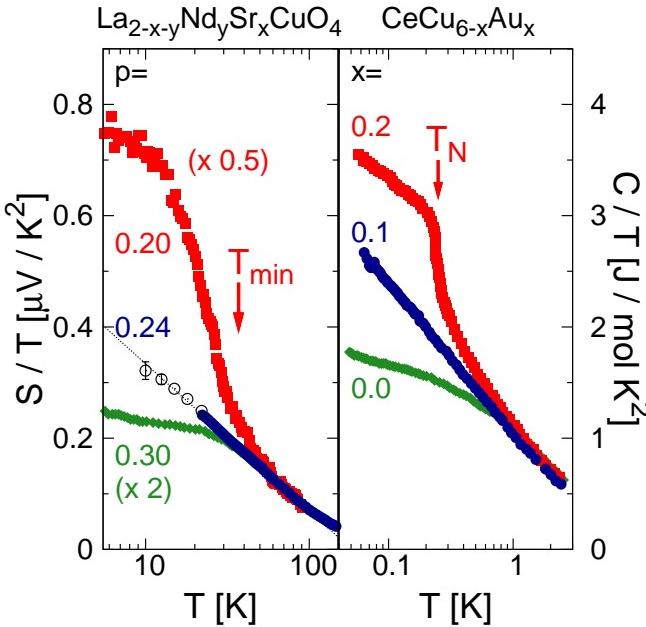


FIG. 3: (Color online) Left panel: Thermopower of  $\text{La}_{2-x-y}\text{Nd}_y\text{Sr}_x\text{CuO}_4$  with  $p = 0.20$  ( $\times 0.5$ ) and  $p = 0.24$  ( $y = 0.4$ , this work), compared to that of LSCO with  $p = 0.30$  ( $\times 2$ ;  $y = 0$ , from [23]), plotted as  $S/T$  vs  $\log T$ . Right panel: Specific heat of the heavy-fermion metal  $\text{CeCu}_{6-x}\text{Au}_x$  at  $x = 0.0, 0.1$  and  $0.3$ , plotted as  $C_e/T$  vs  $\log T$ , showing the evolution across the quantum critical point at  $x = x_c = 0.1$  where the ordering temperature  $T_N$  goes to zero (from [22]).

been invoked to explain the temperature dependence in Bi-2212 [19], it is not satisfactory for the case of Nd-LSCO and YBCO where neither electron-phonon nor mass-enhancement mechanisms are adequate [20, 21]. Here we propose an electronic origin for both the temperature and doping dependence of  $S$ , at least below 100 K. This is strongly supported by the similarity found in resistivity and Hall effect.

At a doping  $p = 0.24$ , close to  $p^*$ ,  $S/T$  in zero magnetic field shows a perfect  $\ln(T_0/T)$  dependence from 100 K down to  $T_c$ . Application of a magnetic field  $H \parallel c = 15$  T to push  $T_c$  down is seen to slightly suppress  $S/T$  below this  $\ln(T_0/T)$  dependence for  $T < 40$  K. By extrapolating the field dependence of  $S/T$  to  $H = 0$ , as shown in the inset to Fig. 2, we can track the zero-field  $S/T$  at temperatures below  $T_c(H = 0)$ . This shows that the  $\ln(1/T)$  regime persists at least down to 10 K, within the uncertainty of this extrapolation, *i.e.* over a full decade of temperature.

This  $\ln(T_0/T)$  dependence of  $S/T$  is strongly reminiscent of the  $\ln(T_0/T)$  dependence observed in  $C_e/T$  at the quantum critical point of various heavy-fermion metals [13]. In Fig. 3, we compare  $S/T$  in Nd-LSCO with  $C_e/T$  in the heavy-fermion metal  $\text{CeCu}_{6-x}\text{Au}_x$  [22], each at three concentrations: below, at and above their respec-

tive critical points,  $p^*$  and  $x_c$ . By substituting Au in  $\text{CeCu}_6$ , antiferromagnetic order is made to appear beyond a critical concentration  $x = x_c = 0.1$ , with an ordering temperature  $T_N$  that rises linearly with  $x$  [13, 22]. In the absence of data on Nd-LSCO at  $p > p^*$ , we compare with data on LSCO ( $y = 0$ ) at  $x = p = 0.30$  [23]. Given that both materials exhibit virtually identical resistivity and thermopower above  $T^*$  [16], it is reasonable to assume they also do above  $p^*$ .

The similarity is remarkable, both materials displaying the three distinctive regimes of quantum criticality: relatively flat in the Fermi-liquid state, logarithmically divergent at the critical point, and a jump in the ordered state. The characteristic temperature scale  $T_0$  in the  $\ln(T_0/T)$  dependence of either  $S/T$  or  $C_e/T$  is of course vastly different in the two materials, by roughly two orders of magnitude, as are the ordering and pseudogap temperatures,  $T_N$  and  $T^*$ . This qualitative similarity reinforces the case for a quantum phase transition in Nd-LSCO at  $p^*$ , previously made on the basis of resistivity [4], whose three regimes are displayed in Fig. 4: quadratic in the Fermi-liquid state, linear at the critical point, and an upturn below that point.

There is also a strong similarity with the electron doped cuprate  $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}$  (PCCO), where the case for a quantum critical point is well established [25]. In the  $T \rightarrow 0$  limit, both  $R_H$  and  $S/T$  in PCCO show an abrupt change as the doping  $x$  drops below the critical doping  $x_c$ , signalling the change in Fermi surface from a large hole cylinder to a combination of small electron and hole pockets [26, 27]. The two coefficients track each other, as equivalent measures of the effective carrier density [26]. At  $x = x_c$ ,  $\rho(T)$  is again linear in temperature at low  $T$  [28]. These typical signatures of a quantum critical point have been attributed to the loss of antiferromagnetic order near  $x_c$  [29], and the quantum fluctuations thereof.

In a model of charge carriers on a three-dimensional Fermi surface scattered by two-dimensional antiferromagnetic spin fluctuations, transport properties near the magnetic quantum critical point are found to be dominated by “hot spots”, points on the Fermi surface connected by the ordering wavevector. In this case, calculations show that  $\rho(T) \propto T$ ,  $C_e/T \propto \ln(T_0/T)$  and  $S/T \propto \ln(T_0/T)$ , where  $k_B T_0$  is an energy scale on the order of the bandwidth [30]. This naturally accounts for the different temperature scales observed in Nd-LSCO and  $\text{CeCu}_{6-x}\text{Au}_x$  where  $T_0 \simeq 170$  K in the former and 4 K in the latter, since the Fermi velocity is about  $10^5$  m/s in cuprates and  $10^3$  m/s in heavy-fermion metals.

The strong empirical similarity with both heavy-fermion metals and electron-doped cuprates makes a compelling case for a quantum critical point at  $p^*$  in Nd-LSCO. The nature of the order below  $p^*$  seems to involve both spin and charge degrees of freedom. On the one hand, superlattice Bragg peaks observed in Nd-

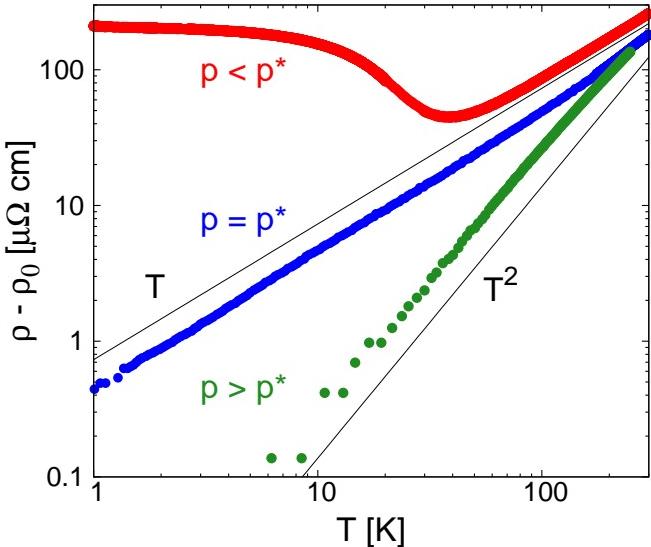


FIG. 4: (Color online) Temperature dependent part of the resistivity,  $\rho(T) - \rho_0$ , vs  $\log T$  for  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  with  $p = 0.20$  ( $p < p^*$ ) and  $p = 0.24$  ( $p = p^*$ ), from [4], compared to that of LSCO with  $p = 0.30$  ( $p > p^*$ ), from [24].  $\rho_0$  is the value to which  $\rho(T)$  extrapolates at  $T = 0$ ; for Nd-LSCO at  $p = 0.20$ , the extrapolation is based only on data above  $T^* = 80$  K.

LSCO by neutron diffraction show that a static (or slow) spin modulation at low temperature persists all the way up to  $p \approx p^*$  [6]. On the other hand, the upturn in  $\rho(T)$  at  $T_{\min}$  coincides with the onset of charge order [4], which occurs at a temperature somewhat above the onset of spin modulation [6]. In other words, the pseudogap phase below  $T^*$  (and  $p^*$ ) appears to be a phase with “stripe” order, perhaps short-range or fluctuating above  $T_{\min}$ .

The impact of stripe order on the Fermi surface of a hole-doped cuprate has been calculated [31]. The large hole-like cylinder is found to reconstruct in a way that depends on the strength of the spin and charge potentials. Calculations of the associated Hall coefficient predict a rise in  $R_H$  with the onset of charge order [32], as observed experimentally in Nd-LSCO when going from  $p = 0.24$  to  $p = 0.20$  [4]. Spin order can cause a drop in  $R_H$ , which can even become negative [32], as a result of an electron pocket being present in the reconstructed Fermi surface [31]. Such a drop is indeed seen in Nd-LSCO at lower doping, in the vicinity of  $p = 1/8$ , where  $R_H(T \rightarrow 0) \approx 0$  [16]. The fact that a large drop in  $R_H(T)$  also occurs in YBCO near  $p = 1/8$  [3], starting at a very similar temperature, points to a common underlying cause of Fermi-surface reconstruction.

In conclusion, the combination of resistivity, Hall coefficient and thermopower in Nd-LSCO makes a compelling case that the pseudogap phase in this high- $T_c$  superconductor ends at a quantum critical point located inside the superconducting dome at  $p \approx 0.24$ . All three transport coefficients undergo a simultaneous rise below

a temperature  $T_{\min}$  which coincides with the onset of charge order seen by other probes. This strongly suggests that the Fermi surface is reconstructed by “stripe” order. Given that a linear- $T$  resistivity is a universal property of cuprates near optimal doping, it is likely that a common mechanism is at play, associated with such a quantum critical point, in analogy with heavy-fermion metals.

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